

Software development for continuous-gas-flow AMS

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Abstract: The National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) Facility at Woods Hole Oceanographic Institution is presently completing installation of a novel continuous-flow AMS system. A multi-year development of an AMS microwave gas ion source in collaboration with Atomic Energy Canada Limited (AECL), Chalk River, has preceded this final step of an implementation that is expected to add a new dimension to ^{14}C AMS. National Instruments, NIM, and CAMAC modules have been programmed with LabVIEW on a Windows XP platform to form the basis for data acquisition. In this paper we discuss possible applications and include simulations of expected data acquisition scenarios like real-time AMS analysis of chromatograms. Particular attention is given to issues of synchronization between rapidly changing input amplitudes and signal processing cycles in hardware and software.

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Introduction

A new 500 kV AMS system[1] at the National Ocean Sciences AMS Facility, designed to measure ^{14}C in a continuously flowing gas stream (CFAMS), is reaching its final stage of assembly with the impending completion of our microwave gas ion source, designed in collaboration with John Wills and his colleagues from AECL, Chalk River [2-4]. We have obtained data with the proto-type ion source on our original 3MV Tandatron and used it to model the expected performance of the new system under various conditions.

^{14}C data acquisition system

One of the important requirements of a continuous-flow AMS system is minimal dead time in the ^{14}C acquisition to enable the analysis of rapidly succeeding CO_2 pulses in real time. The CFAMS sequential injector operates at ~ 9.7 Hz (1.2 ms ^{12}C , 2.4 ms ^{13}C , 100 ms ^{14}C) where the control voltage pattern to the Trek high-voltage bouncer supply is generated by a National Instruments PXI 6251 DAQ / 6602 Timing I/O board combination programmed in PC-based LabView. Analog to digital conversion of the Faraday cup signals (for high energy ^{12}C and ^{13}C Faraday cups, as well as a low energy Faraday cup) is measured on these same boards and synchronized with the bouncer supply control waveform, with controlled delays to allow for beam tracking and electrometer settling time. A gate voltage is generated by the DAC/ADC boards with a built-in adjustable delay for control of the ^{14}C counting electronics.

To achieve largely dead time free acquisition we use a 5 MB/s SCSI interfaced CAMAC setup with an ORTEC AD413A 8k ADC and a CMC203 Fera Driver with 1 MWord FIFO buffer[5], gated within the ^{14}C bouncer cycle. At rates below 1 kHz the measured dead time is less than 3 ms. To account for dead time at high count rates a Tennelec TC814 90 Hz pulser signal is fed into the pre-amplifier together with the ^{14}C surface barrier detector signals, and acquired in the high-channel section of the ADC. Directly counted pulser signals are compared to the pulser peak in the ADC spectrum for dead time correction. This LabView controlled acquisition system has proven reliable under most circumstances during standard sputter ion source operation.

CO₂ injection systems

There are a number of developments underway to design sources of continuously flowing CO₂ gas (chromatography, laser ablation, controlled combustion, and hydrolysis). All of these methods will have to comply with one limitation: the desirable flow rate into the gas ion source is ~3 $\mu\text{l/s}$. Higher or much lower rates (in absence of a make-up carrier gas) will lead to vacuum disruption or quenching of the microwave plasma, respectively. To ensure constant flow at this rate we have applied an open split method (commonly used in stable isotope mass spectrometry): a capillary is cut to the length effecting flow rates of ~3 $\mu\text{l/s}$ at 1 atm head pressure. A larger I.D. capillary from the gas supply is sleeved over the end of the first capillary but not sealed gas tight. Source gas at >1 atm will be split at this insertion point, allowing excess gas to escape. Using this method we generated square injection pulses of 30 s duration and established a system response function. Of particular interest is the pulse decay time as it affects the memory of the system between injected pulses. It appears that there are at least two decay time constants in the tail of the pulses. We fitted the data with a third order logarithmic function that describes the tail of the ¹²C current pulse well (see fig. 1). In the following we will use this function to predict the response of the AMS system to other types of pulses.

Expected ¹⁴C response

An injected square pulse of 30 s duration results in the ¹⁴C spectrum in figure 2 that was simulated by assuming statistical noise and folding the data with the fitted tail function up to 24 s after each ¹⁴C event. The simulation very closely resembles data taken with the prototype ion source on the Tandetron system. We measured the efficiency of the new

CFAMS system (with MC-SNICS sputter source) to be ~4.5%, over four times larger than that of the Tandetron system with its hemispherical ionizer sputter source. With the gas ion source the system efficiency of the Tandetron was determined to be 0.07%. Therefore we expect that value to be at least 0.15% for CFAMS. The 100 μ l square pulse (~50 μ g C) then would generate ~5000 ^{14}C counts as shown here.

Gas Chromatography

One of the most intriguing aspects of this development is the possibility to analyze ^{14}C in the effluent from a chromatograph after in-line combustion. As a test of the expected AMS response to a stream of chromatogram peaks from a megabore GC column we took a commonly-used standard in chromatography, fatty acid methyl ester (FAME) and assumed that the combustion effluent is reasonably well represented by the GC spectrum. Further it was assumed that the larger of the peaks in the spectrum contained ~20 nmol C. Again folding the tail function into the data as described before, the resulting AMS ^{14}C spectrum is shown in figure 3 for a small section of the FAME spectrum. Clearly, there is no problem resolving larger and reasonably well separated peaks. For this simulation the fraction modern carbon (fmc) content was assumed to be 1 for 18:0 and 18:2. As an illustration of the effect of ^{14}C enriched compounds, 18:1 was assumed to have fmc = 10.

Programmed temperature pyrolysis / combustion

Another development in our laboratory concerns a method to separate classes of compounds in a sample by temperature-controlled pyrolysis and combustion [6]. A schematic of the setup is shown in figure 4. Again the effluent of this device is fed

through a capillary into the gas ion source. The input and expected ^{14}C response is displayed in figure 5. Here, the separation of individual components of the spectrum and the interpretation of the results will be more challenging than for a GC spectrum. In this example a mixture of diesel soot and apple leaves was combusted under steadily rising temperature to distinguish the labile modern compounds clearly from the refractory soot. As it turns out both components mix at all levels of reactivity, rendering the lead section less modern and the trailing compounds less old. A LabView panel as shown in figure 5 might be used to display the results of a programmed temperature combustion run. This should help investigators to quickly establish the presence or absence of fossil components in an unknown sample, before going to more elaborate methods to separate individual compounds.

Conclusion

The advantage of continuous-flow AMS over traditional methods is that it will be possible to scan CO_2 from unknown materials for presence or absence of ^{14}C in an instant survey. The precision of the results may in some cases be a few percent, enough to classify the material but not enough for an accurate determination. At this point the decision can be made whether or not to continue the AMS analysis. For a chromatogram that may mean repeated runs or isolation of specific fractions for separate measurement. The avoidance of the graphitization step saves valuable time especially in pilot projects where quick estimates are needed to establish the feasibility of a research proposal. The applications discussed here for the new CFAMS system, among others, will surely bring exciting new challenges to the NOSAMS facility. A major challenge will be the

determination of background levels, and ultimately classic nuclear physics peak fitting routines will aid in the interpretation of results. In figure 6 the expected number of ^{14}C counts from a gas sample is plotted against the total mass of the sample in μmol (reflecting a range of 0 – 120 μg of carbon), for fraction modern values of 1, 10, and 100, conservatively assuming a system efficiency of 0.15%.

Acknowledgment

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References

- [1] M.L. Roberts, R.J. Schneider, K.F. von Reden, J.S.C. Wills, B.X. Han, J.M. Hayes, B.E. Rosenheim and W.J. Jenkins, Nucl. Inst. & Meth. in Physics Research B 259 (2007) 83-87.
- [2] R.J. Schneider, S-W. Kim, K.F. von Reden, Hayes, J.M., Wills, J.S.C., V. Griffin, A. Sessions, and S. Sylva, Nucl. Inst. & Meth. in Physics Research B 223-224 (2004) 149-154.
- [3] B.X. Han, K.F. von Reden, M.L. Roberts, R.J. Schneider, J.M. Hayes and W.J. Jenkins, Nucl. Inst. & Meth. in Physics Research B 259 (2007) 111-117.
- [4] K. von Reden, M. Roberts, B. Han, R. Schneider, J. Wills, A.I.P. Conference Proceedings 925 (2007) 341 – 345.
- [5] *Cheesecote Mountain CAMAC* CMC203 Fera Driver/PGA, www.cmcamac.com.
- [6] B. E. Rosenheim, J. M. Hayes, A. Benthien, M. B. Day, H. Schrum, and E. W. Domack, *Geochemistry Geophysics Geosystems*, Submitted, 2007.

Figure Captions

Figure 1: Trailing edge of a square ^{12}C pulse measured with the prototype gas ion source in the high-energy AMS Faraday cup. The best-fit tail function is used in the simulations of this paper.

Figure 2: Square 100 μl CO_2 pulse and expected ^{14}C counts.

Figure 3: Chromatogram section with single-pass and 10-pass ^{14}C simulations. See text for details.

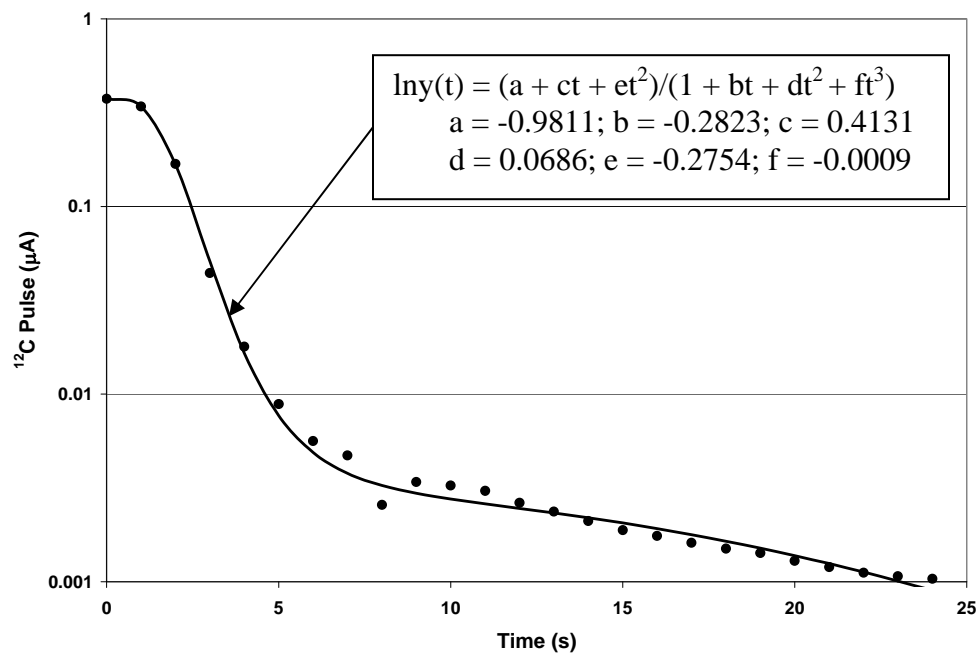
Figure 4 (from [6]): Schematic of a programmable temperature combustion/pyrolysis setup as used in our laboratory.

Figure 5: LabView screen of a PTC/S run with simulated single-pass and 10-pass ^{14}C results from a mix of modern and fossil carbon sources.

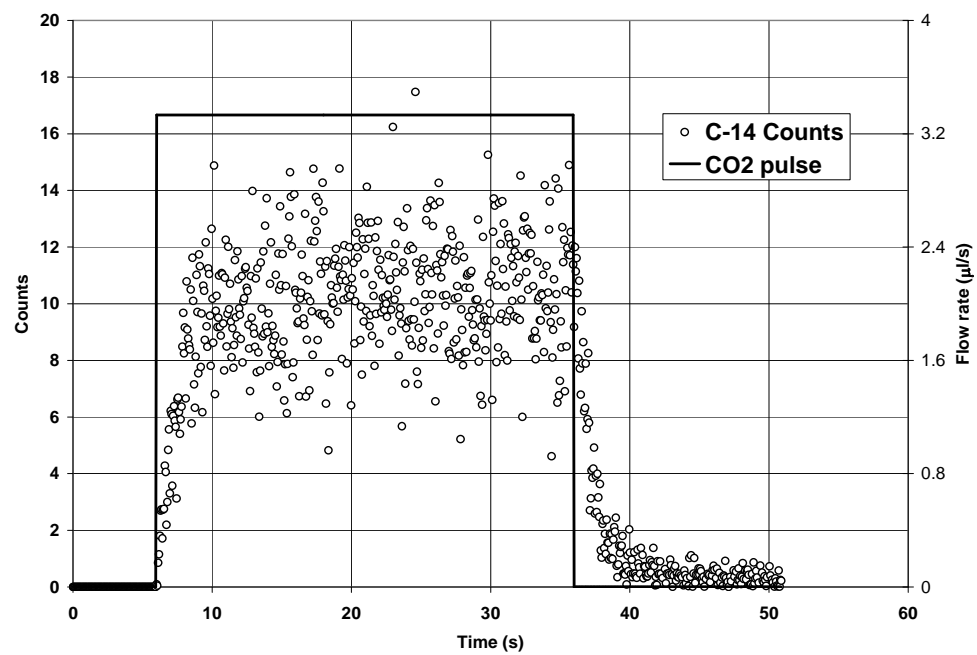
Figure 6: Expected ^{14}C counts in the CFAMS system as function of sample mass. The overall system efficiency was conservatively assumed to be 0.15%.

Figures

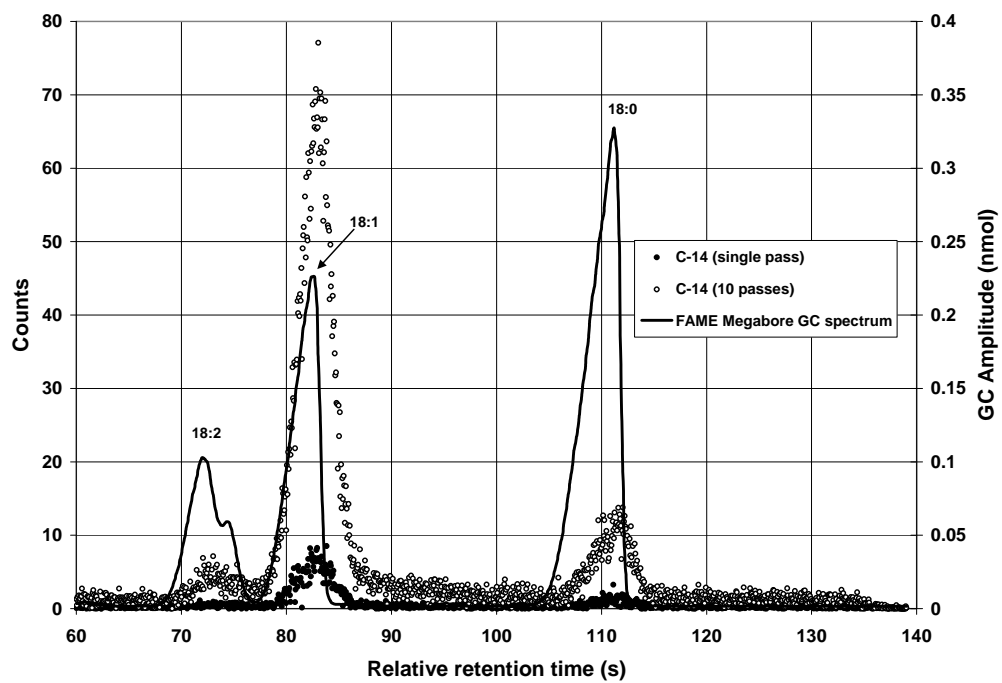
(Fig. 1)



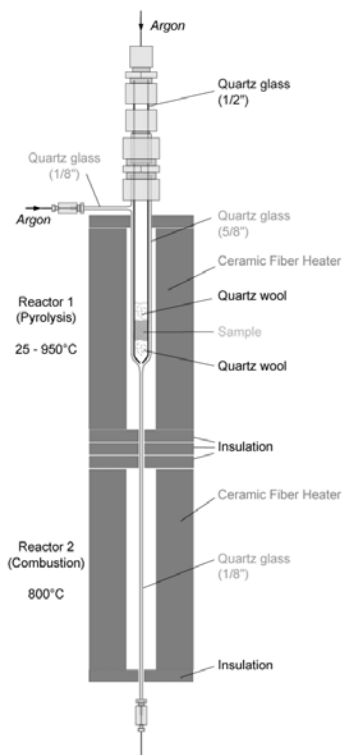
(Fig. 2)



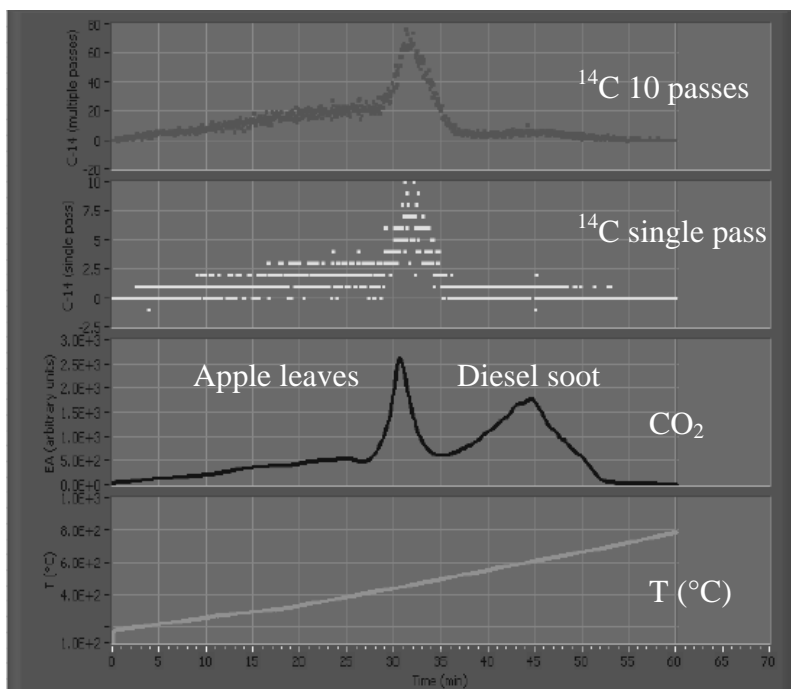
(Fig. 3)



(Fig. 4)



(Fig. 5)



(Fig. 6)

